Southern Fine Particulate Monitoring Project

Seventh Quarterly Progress Report

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Abstract

This quarterly report presents results and analysis of continuous onsite ambient fine particulate data at the North Birmingham sampling site during the April - June, 2002 study period. The continuous data include $PM_{2.5}$ mass concentrations measured by TEOM, particle sulfate using the R&P 8400S monitor, particle size distributions measured by SMPS and APS monitors, and $PM_{2.5}$ light scattering extinction coefficient as measured by nephelometer. Some performance indications for the upgraded APS model 3321 are described in the report. Clear improvement of particle size measurements in the submicron range were noted. Coarse particle measurement with the APS appears to be somewhat improved as well, but particle loss effects are still significant for this size range. During the quarter preliminary data analysis and modeling studies were continued to test the potential of the North Birmingham site data for source attribution analyses. We anticipate that these analyses will provide good separations of the effects of major source classes and spatial source clusters, and will provide useful information relevant to $PM_{2.5}$ implementation strategies.

Table of Contents

List of Figures

Introduction

This is the seventh quarterly progress report of the "Southern Fine Particulate Monitoring Project", funded by the U.S. Department of Energy's National Energy Technology Laboratory under DOE Cooperative Agreement No. DE-FC26-00NT40770 to Southern Research Institute (SRI). In this two year project SRI will conduct detailed studies of ambient fine particulate matter in the Birmingham, AL metropolitan area. Project objectives include:

$ Augment existing measurements of primary and secondary aerosols at an established urban southeastern monitoring site
$ Make a detailed database of near-continuous measurements of the time variation of fine particulate mass, composition, and key properties (including particle size distribution)
$ Apply the measurements to source attribution, time/transport properties of fine PM, and implications for management strategies for $PM_{2.5}$
$ Validate and compare key measurement methods used in this study for applicability within other $PM_{2.5}$ research by DOE-FE, EPA, NARSTO, and others.

Summary of Technical Progress

Progress and Plans

During the seventh project quarter, continuous onsite ambient data were collected and monitored. Details include:

$ Prepared July data in general format for modeling study
$ Comparison of results for the original TSI APS 3320 to the upgraded APS 3321
$ Continued monitoring with TEOM, particle sizing instruments, R&P 8400 Sulfate monitor, Radiance M903 Nephelometer, and 43CTL Sulfur Dioxide analyzer
$ Continued Source Attribution analysis using PMF techniques
$ Preliminary results presented at DOE - sponsored *$PM_{2.5}$ and Electric Power Generation Conference,* April 9-10, 2002 in Pittsburgh, PA.

Plans for next quarter include the following:

$ Complete onsite monitoring with continuous monitoring instruments
$ Continue analysis of continuous particulate data
$ Continue source attribution analysis using wind trajectories, receptor methods
$ Complete and submit project final report.

Problems and Assessment for Future Progress

During most of the month of April the SMPS was unavailable. In early April the CPC detector began to show error flags, and the laser was found to need replacement. The device was repaired by TSI, returned, tested, and replaced in the shelter on April 26, resulting in three weeks lost SMPS data. As reported last quarter, a replacement computer motherboard was installed in

February. The new computer has been subject to several crashes since it was put into place, resulting in periodic loss of data. The particle sizing instruments shut down immediately in the computer outages, and thus incur the greatest data loss. Data loss from the other devices occurs only when the computer is down more than a day before it is discovered and restarted. This also happened a few times during the quarter, resulting in some TEOM data loss.

Experimental

As reported last quarter, the APS 3320 was sent to TSI to be upgraded to an APS 3321 January 23. According to TSI, the upgrade enhancements provide greater small-particle sizing efficiency, improved accuracy of mass-weighted distributions, and virtual elimination of false background counts. After return and some limited lab testing, the APS and SMPS were reinstalled at the North Birmingham monitoring station March 14, but comparative data were compromised by failure of the SMPS condensation particle counter. After both devices were returned to operation in late April, we were finally able to make a field assessment of the comparative performance of the upgraded APS 3321.

Results and Discussion

Hourly averages of the continuous particulate measurements during the quarter are presented in Figures 1 - 6. The data are plotted together for a meaningful comparison between instruments and data sets. The figures contain the $PM_{2.5}$ mass concentrations measured by the TEOM, 8400S sulfate monitor and integrated size fractions measured by the particle sizing devices. Included are total (submicron) mass concentration as derived from the SMPS measurements, and integrated mass concentrations in the 1 - 2.5 and 2.5 - 10 μm size ranges from the APS measurement data. The $PM_{2.5}$ light scattering extinction coefficient as measured by the M903 nephelometer is plotted on the second Y-axis. In addition, hourly average PM_{10} concentration data were obtained from the Jefferson County Health Department as measured by the county TEOM monitor at the site.

Figures 1 - 3 display the variables associated with the measurements in the fine particulate region. The data sets displayed are the $PM_{2.5}$ TEOM, SMPS total concentration, the 1 - 2.5 μm APS fraction, the 8400S sulfate monitor and the M903 nephelometer. The figures present the same five variables over the months January, February, March, respectively. Figures 4 - 6 represent the variables associated with particulate measurements in the coarse size region, including the PM_{10} TEOM and the 2.5 – 10 μm APS fraction, as well as the $PM_{2.5}$ TEOM concentrations for reference.

<u>APS Comparisons</u>

As described above, the APS 3320 was sent to TSI to be upgraded to an APS 3321 January 23 and reinstalled at the site in mid February after laboratory characterization experiments. Some promising changes in instrument performance were immediately noted. The dead time in the upgraded 3321 was reduced by three orders of magnitude (from around 67,000 units to 65) from its predecessor, promising reduced coincidence loss and better response. The Event 4 parameter, indicating "phantom" large particles, was similarly cut by three orders of magnitude. More

significantly, the count rate corresponding to submicron particles immediately increased by roughly a factor of 5. Since one of the promises of the upgrade enhancements was to provide greater small-particle sizing efficiency, we were eager to obtain more detailed comparison data for indications that these improvements were realized. Unfortunately, the SMPS was removed shortly afterward, and quickly developed repair problems, so only limited field comparisons of the two sizing instruments were possible until late April. As indicated below, the encouraging preliminary indications reported last quarter appear to be verified in the current quarter data.

Figures 7 and 8 present a comparison of integrated mass concentrations measured by the APS 3321 to those measured by the SMPS for their common size region between 0.5 and 1 μm. On the second axis for reference purposes are plotted the ratio of the APS to SMPS results (expressed as a percentage), and the total $PM_{2.5}$ mass concentration as measured by TEOM. As seen in the comparable figure in the last quarterly report, the measurements of the upgraded APS 3321 are of comparable magnitude to the SMPS, but show considerable relative variability. Figure 7 shows a progression of periods during May in which the APS generally reads higher than the SMPS by factors of 1-3, interspersed with periods (May 13-16, 20-25) in which the reverse is true. As Figure 8 continues the comparison into June, a period of general comparability up to June 4 is followed by a general increase in the ratio favoring the APS. The relative sensitivity of the two devices can change rapidly as well, as seen by different responses to mass concentration "peaks" on days such as May 10 and 24. While there is a suggestion that mass concentration "peaks" sometimes shift the ratio in favor of the SMPS, there is no obvious overall correlation with concentration. It is clear that some additional parameter, which changes over the timeframe studied, is driving the relative sensitivity of the two devices. We expect any comparison of the two devices over this size range to be especially sensitive to the detailed size distribution, since the counting efficiency of the APS falls off at smaller sizes, while that of the SMPS decreases near 1 μm. Changes in particle density (which may vary if the composition of the particles varies) will also scale differently for the two instruments.

A scatter plot of comparable hourly average integrated mass concentration data from the two devices is shown in Figure 9. Data from three periods are labeled separately, and regression results for each are shown in the figure. The mean APS to SMPS ratio over the month period is 75 percent, with very poor overall correlation. In the more limited time periods the correlation improves, but the ratio varies over an order of magnitude. The figure clearly shows subsets of highly correlated clusters within the overall scatter. For example, in the 5/1-13 data are clusters with mean slopes of roughly 1.9 and 3, corresponding to the center and "wings", respectively, of the high concentration event of 5/4-7. Another cluster of comparable slope is seen in the second period (5/13-20) data, corresponding to data from 5/17. This cluster occurs immediately after the major cluster of very low slope data from 5/15-16. Clearly these clusters represent periods of more homogeneous particle composition for which the correlation of the two instruments is high. Inspection of the detailed size distributions during these periods confirm that these periods do indeed correspond to periods of changes in the size populations adjacent to the overlap range (that is, either near 0.5 or 1 μm).

Another particle size range of importance is the coarse particle fraction, defined here as 2.5 – 10μm aerodynamic diameter. This fraction lies within the operating range of the APS, although transport losses and sampling biases can affect the accuracy of the measurements for the larger

sizes. To assess the accuracy of the integrated APS size distribution for Coarse PM, hourly averages of this APS fraction were compared to Coarse PM concentrations calculated from the difference of Jefferson County PM_{10} TEOM and our $PM_{2.5}$ TEOM. Figure 10 shows this comparison for a period during this quarter. On the second axis for reference purposes are plotted the individual PM_{10} and $PM_{2.5}$ TEOM mass concentrations used to construct the calculated Coarse PM concentrations.

Several caveats should be mentioned regarding this use of the TEOM measurements. First, since it is a difference calculation, the relative accuracy of the calculated Coarse PM concentrations will be poor when the difference is small. As can be seen from the upper traces, this does in fact occur for much of the study period, since most of the ambient PM is in fact submicron and thus should contribute equally to both TEOM samplers. Significantly, the Jefferson County PM_{10} TEOM operates using the original 50°C protocol, while our $PM_{2.5}$ TEOM operates at 30°C using the R&P SES Nafion drier. The effect of this should be to reduce the PM_{10} TEOM values in the presence of semivolatile organic and nitrate, resulting in a probable low bias of unknown magnitude in the calculated Coarse PM concentrations. Some indication of these effects is seen in the presence of a few negative calculated Coarse PM concentrations. A further complication is that while the TEOM samplers are both onsite, they are in different shelters, so a few periods of transient local influences can be seen in the plot.

In Figure 10, as was reported earlier for the original APS, we see that the APS coarse fraction generally tracks the calculated TEOM Coarse PM concentrations but is lower in magnitude. This effect can be better illustrated in the pairwise scatter plot of the two measurements in Figure 11. Within the scatter band of the measurements, the APS coarse readings are roughly a third of the corresponding calculated TEOM values. In spite of the scatter, the correlation between the values is much better than that of Figure 9, and shows no sign of similar systematic effects of other fluctuating parameters. Comparable calculations were made using the original APS 3320 a year earlier and in January just before the upgrade. The mean 35% ratio of the two measurements here is slightly higher than 25% and 20%, respectively, for those periods, with comparable scatter. We attribute the overall lower efficiency to particle losses in the inlet and transport lines leading to the APS. It is not clear that the efficiency differences are due to different APS sensitivity rather than differences in particle transmission losses, although we assume this to be the case since the older APS had been losing sensitivity in other size ranges.

Source Attribution Studies

In the Sixth quarterly report we presented initial results from preliminary data analysis and modeling studies to test the potential of the North Birmingham site data for source attribution analyses. These preliminary results were also presented at DOE - sponsored *$PM_{2.5}$ and Electric Power Generation Conference,* April 9-10, 2002 in Pittsburgh, PA. During the quarter we continued these analyses, focusing on further trajectory analyses of sulfate and SO_2 episodes and receptor analyses using data from the continuous gas and particulate monitors at the site. While these analyses are still in progress, we have confirmed the applicability of continuous gas and particulate monitor data to the source attribution receptor analysis. Discrete size bands from the SMPS and PM coarse measurements have been shown to have predictive strength in these analyses as well as the composition data. The results of these analyses will be presented in the

project final report.

Conclusions

Data collected in this quarter complete a full year of data for the full set of instruments in the current sample shelter. The source attribution studies begun last quarter will be extended and augmented by PMF receptor model analysis using the continuous data sets. We anticipate that these analyses will provide good separations of the effects of major source classes and spatial source clusters, and will provide useful information relevant to $PM_{2.5}$ implementation strategies.

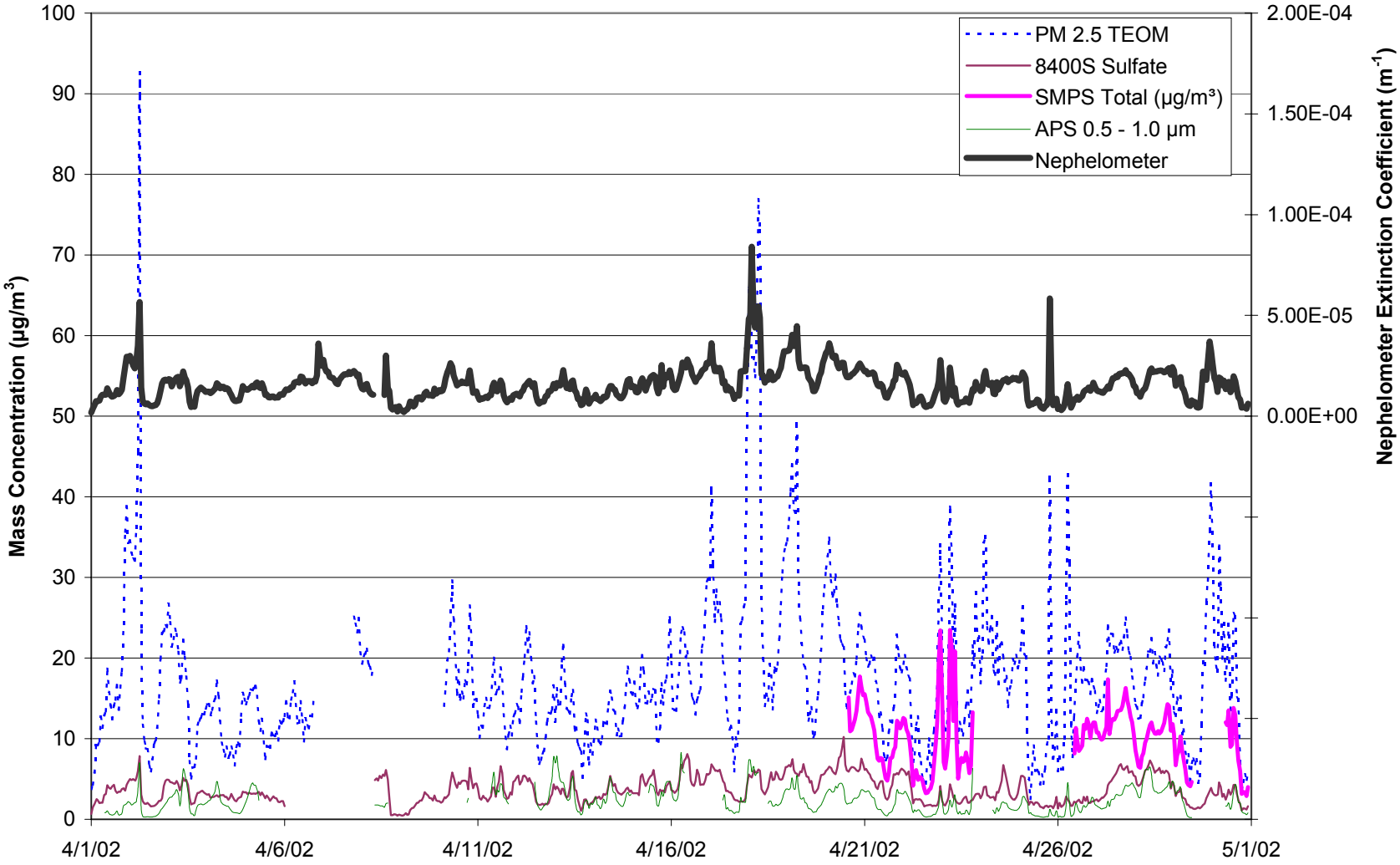

Figure 1. Hourly averaged fine particle data from the North Birmingham site during the period of April 1 – April 30, 2002.

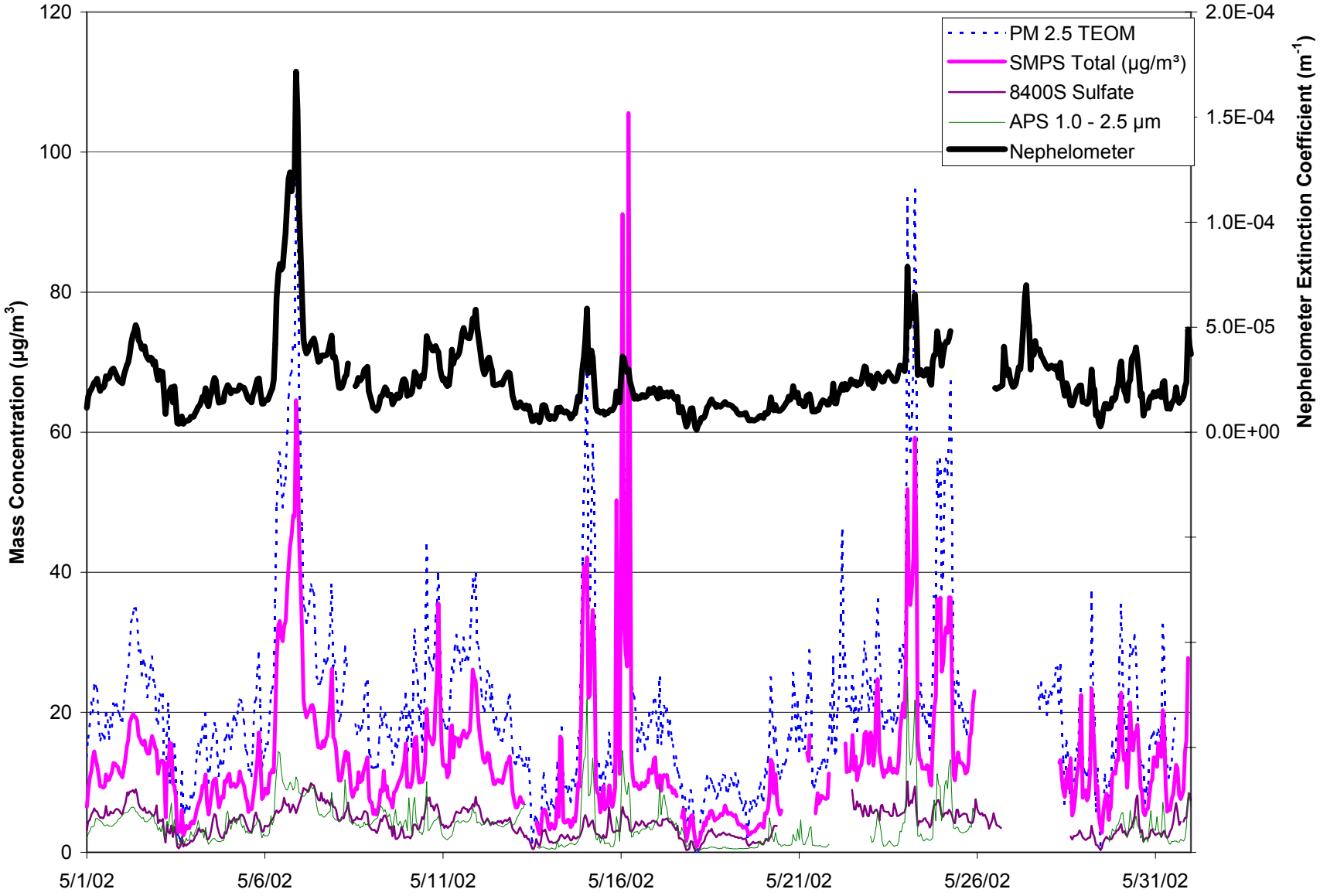

Figure 2. Hourly averaged fine particle data from the North Birmingham site during the period of May 1 – May 31, 2002.

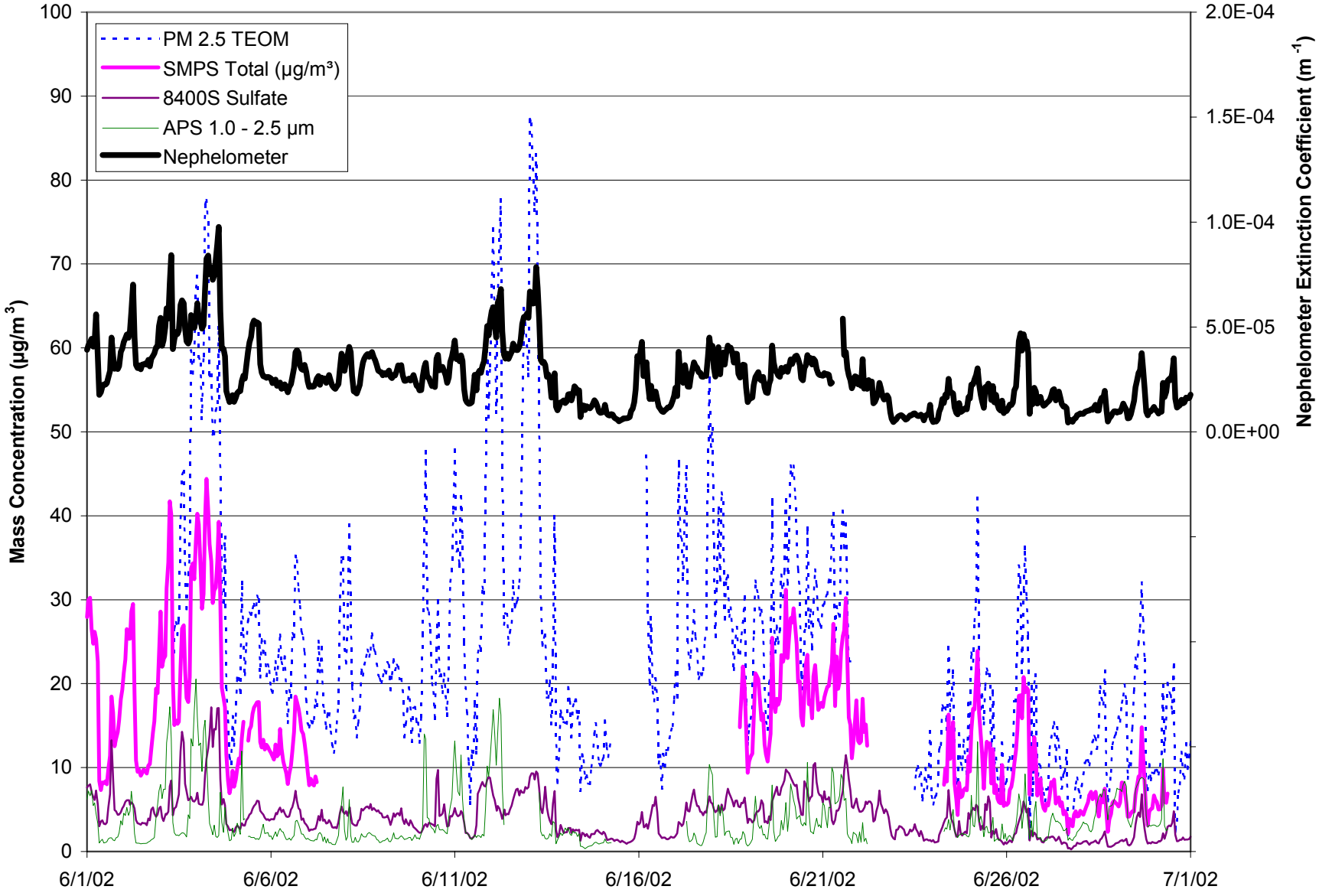

Figure 3. Hourly averaged fine particle data from the North Birmingham site during the period of June 1 – June 30, 2002.

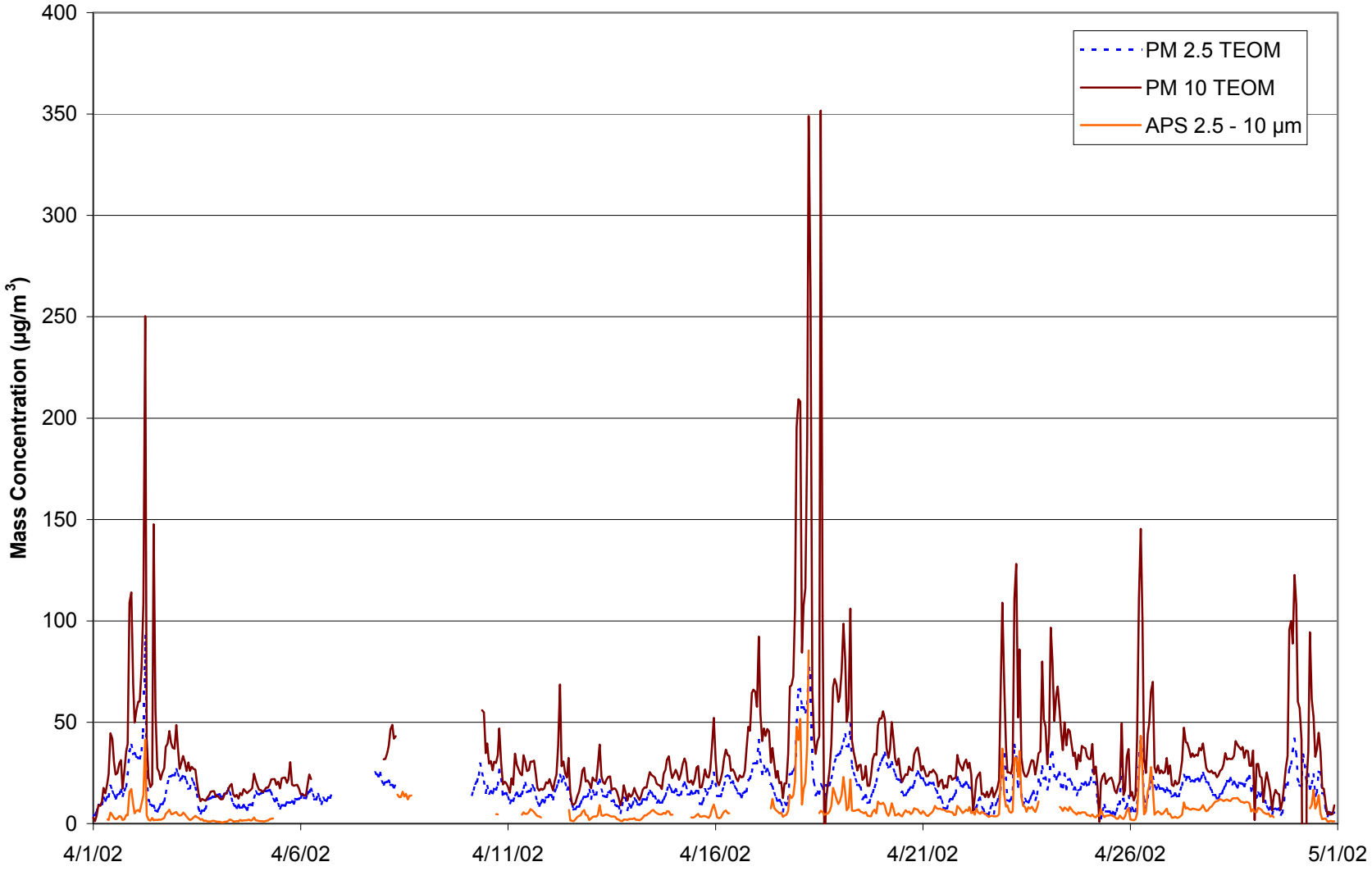

Figure 4. Hourly averaged coarse particle data from the North Birmingham site during the period of April 1 – April 30, 2002. Also included are PM_{10} concentrations reported by Jefferson County.

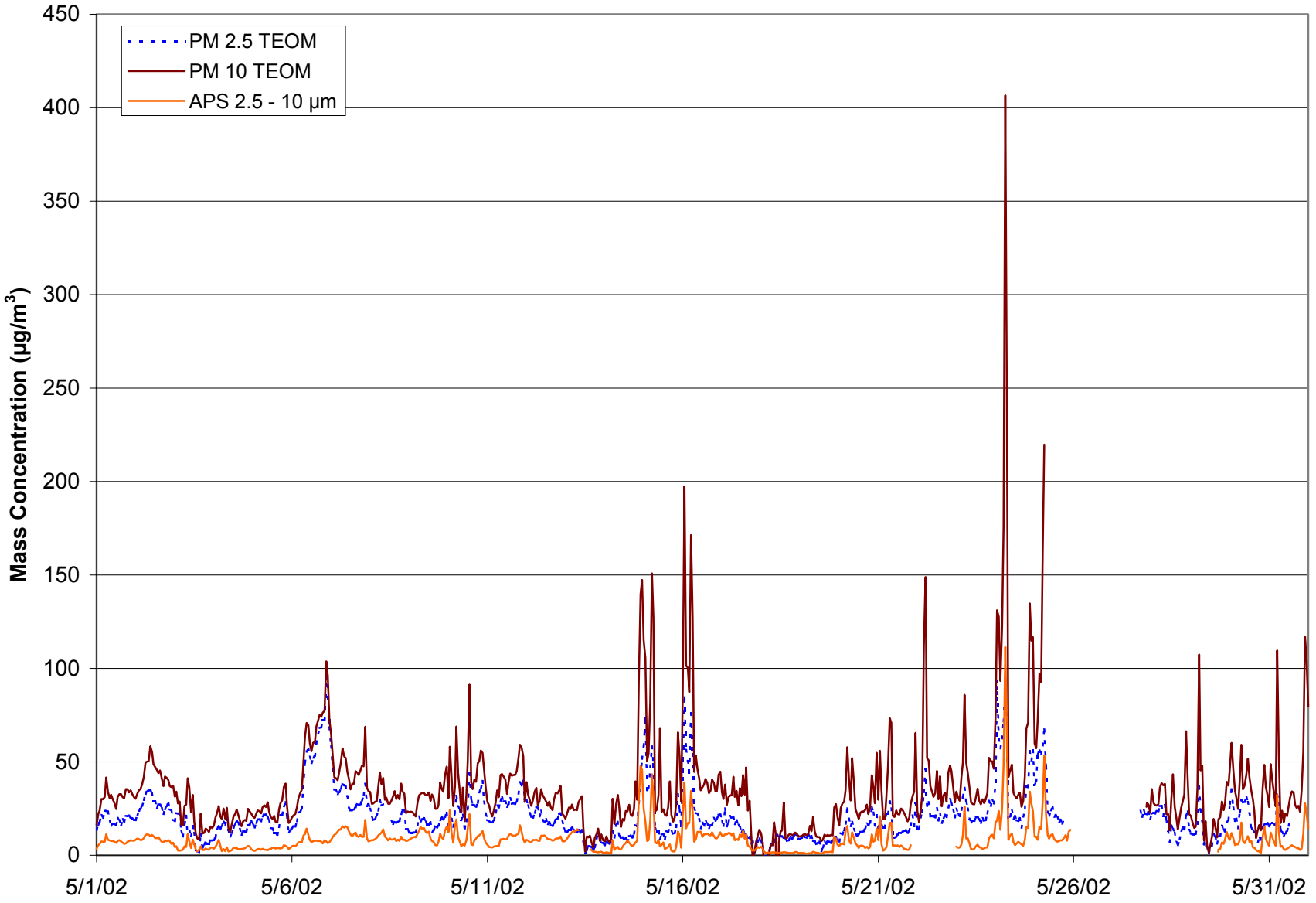

Figure 5. Hourly averaged coarse particle data from the North Birmingham site during the period May 1 – May 31, 2002. Also included are PM_{10} concentrations reported by Jefferson County.

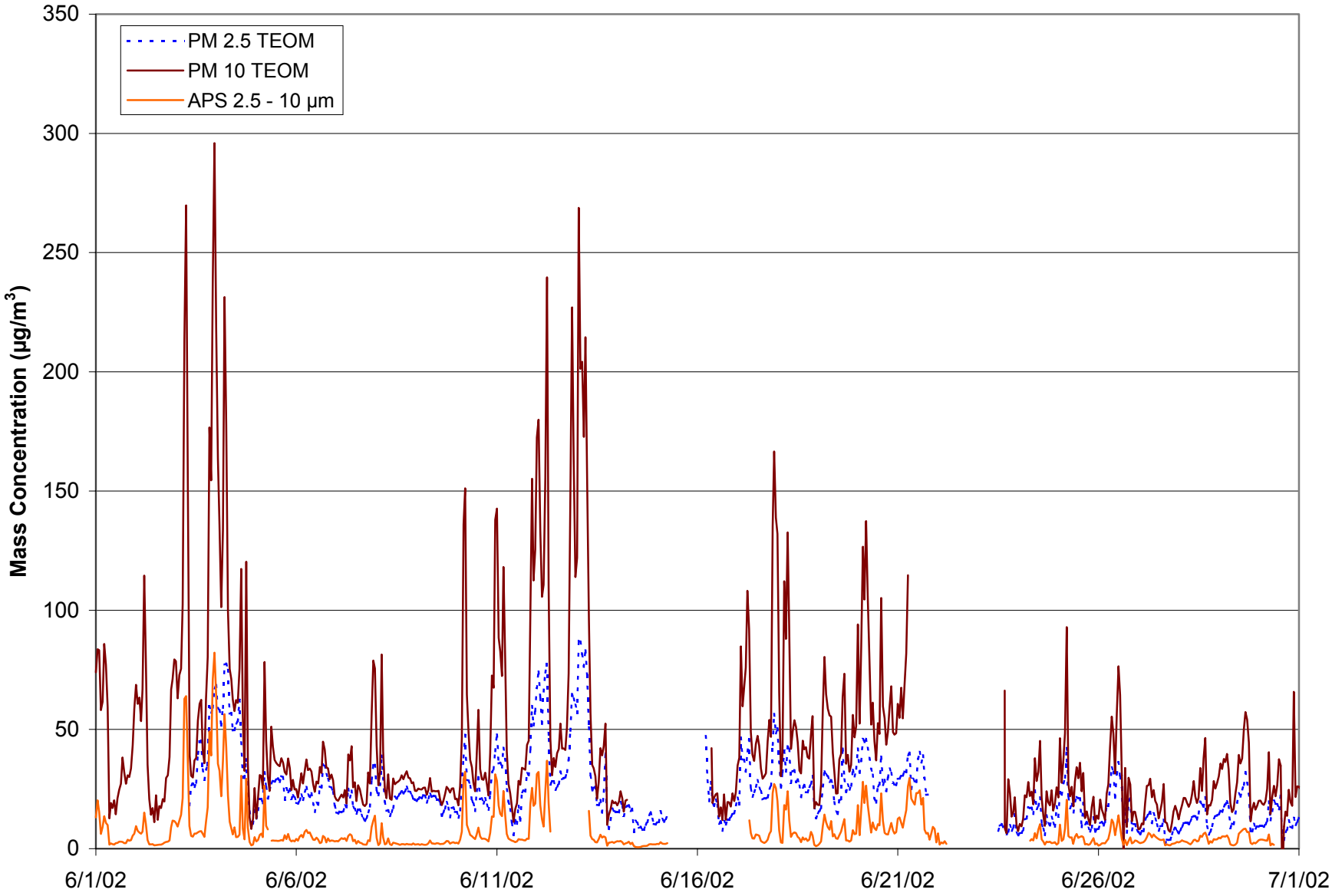

Figure 6. Hourly averaged coarse particle data from the North Birmingham site during the period of June 1 – June 30, 2002. Also included are PM_{10} concentrations reported by Jefferson County.

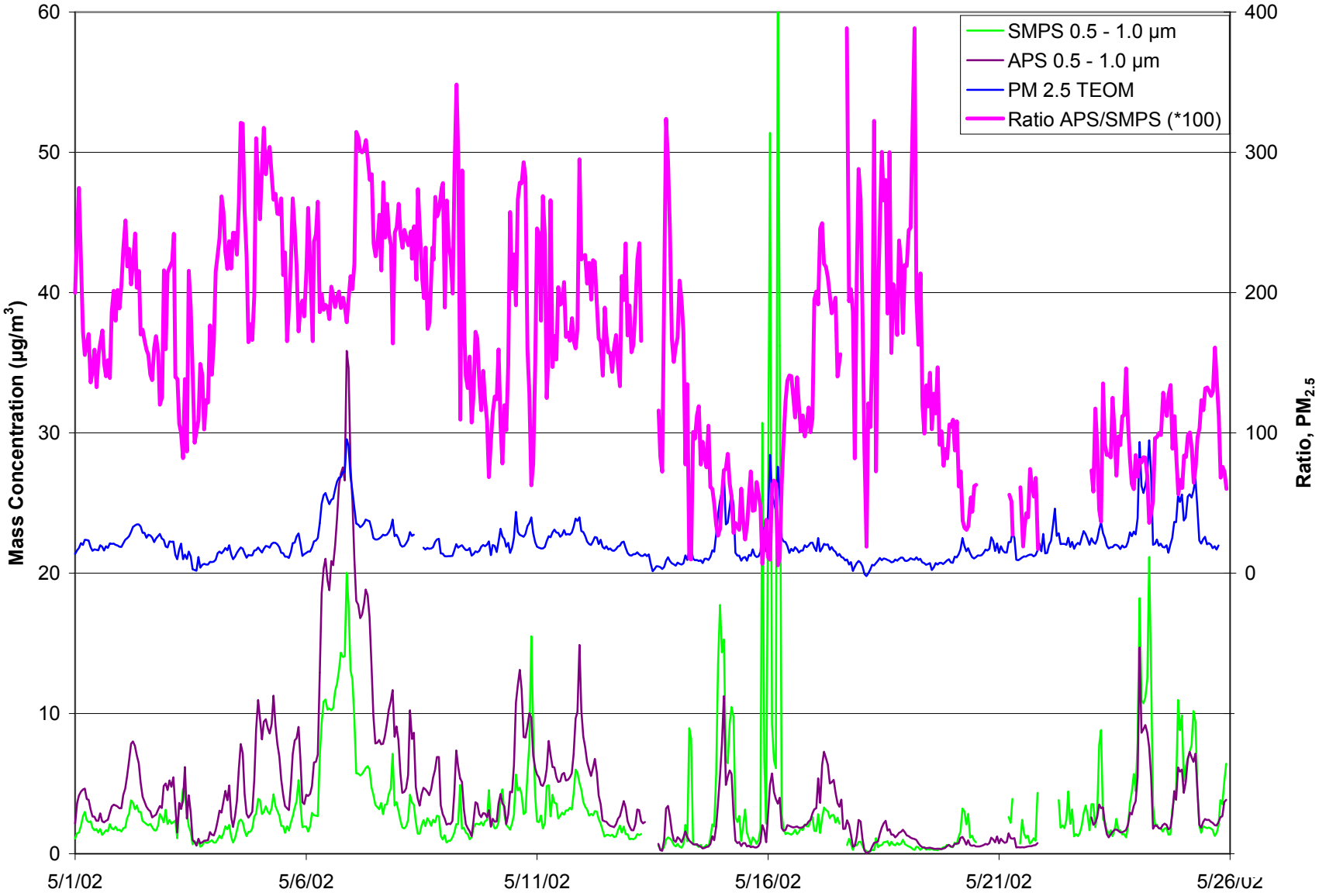

Figure 7. Comparison of the APS 3321 and SMPS 3394 mass concentrations in the overlapping size region of 0.5 - 1.0 µm for May 1 - 26, 2002. The secondary Y-axis presents the ratio of the particle size instrument mass concentrations, and the $PM_{2.5}$ mass concentration.

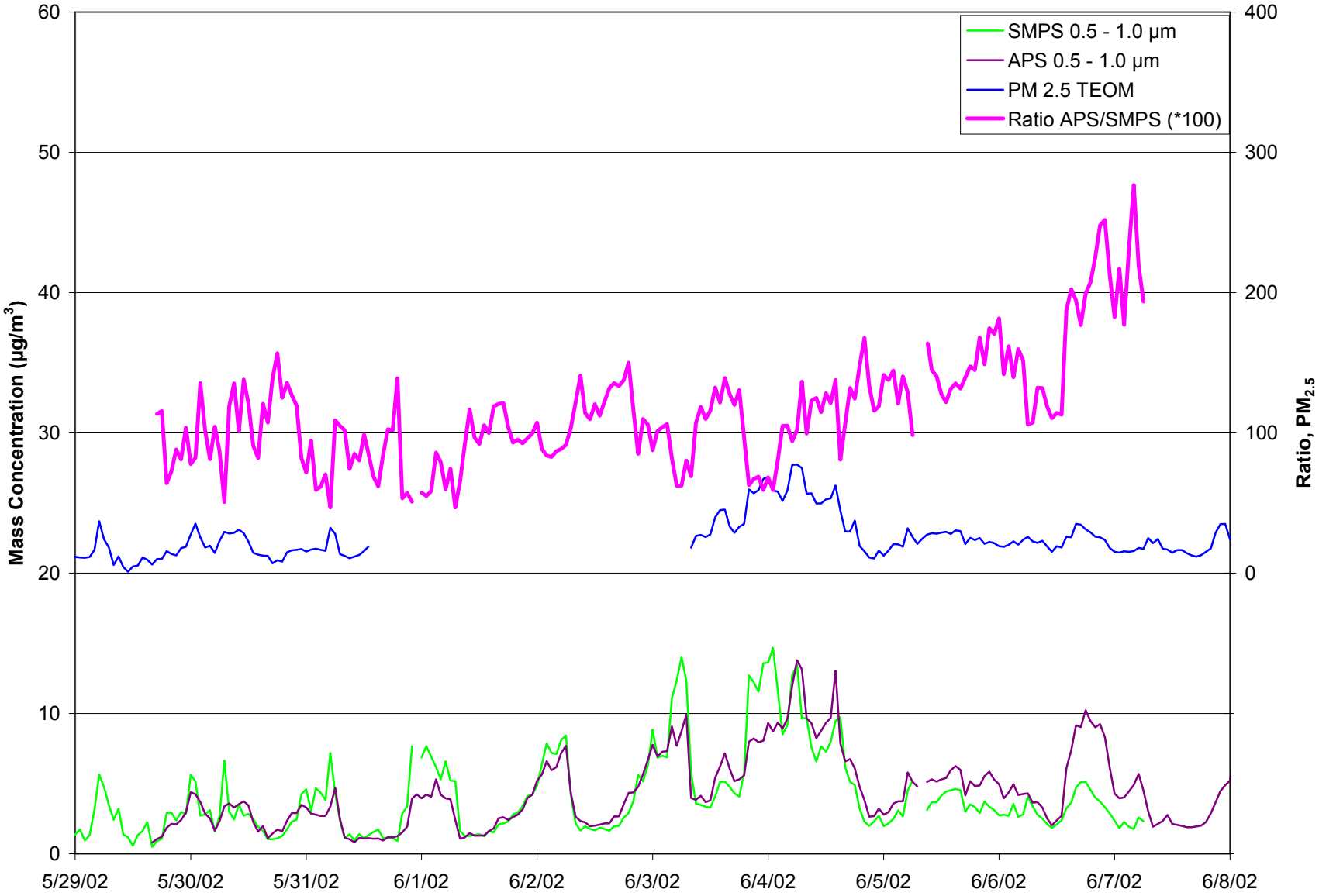

Figure 8. Comparison of the APS 3321 and SMPS 3394 mass concentrations in the overlapping size region of 0.5 - 1.0 µm for May 29 – June 8, 2002. The secondary Y-axis presents the ratio of the particle size instrument mass concentrations, and the $PM_{2.5}$ mass concentration.

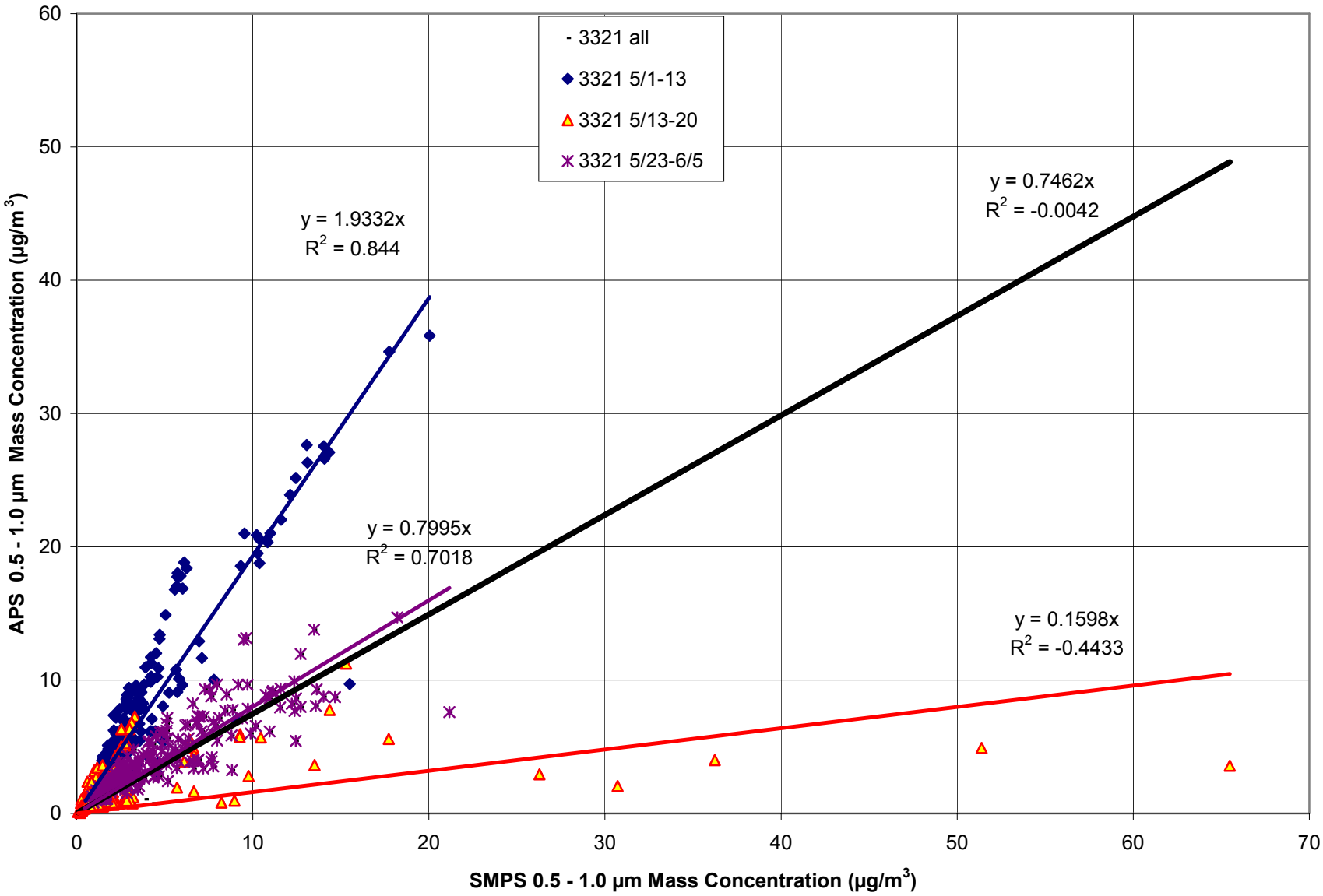

Figure 9. Scatter Plot Comparison of the APS 3321 to the SMPS 3934 concentrations in the 0.5 - 1.0 µm size region for several periods during the quarter.

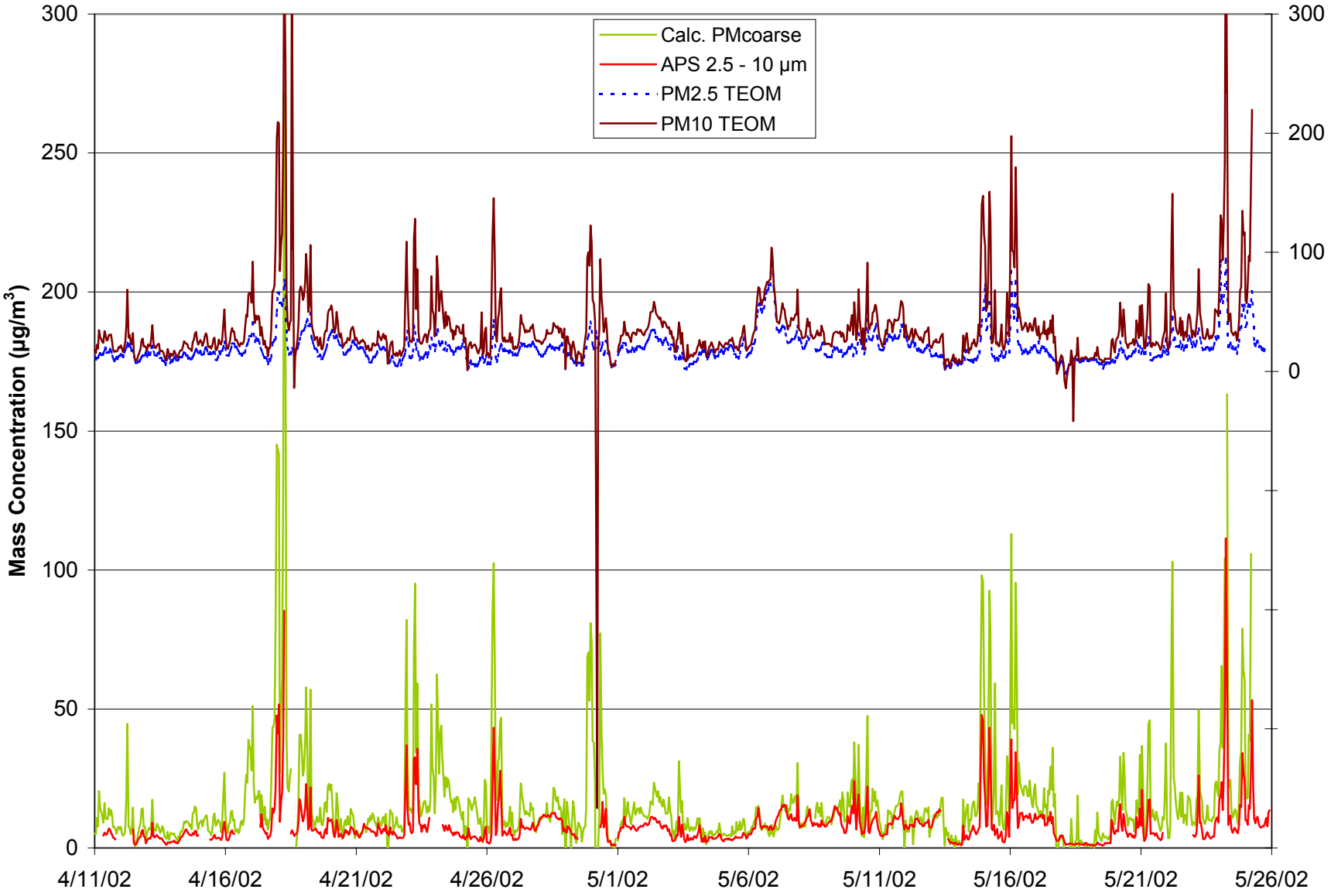

Figure 10. Coarse ($PM_{10} - PM_{2.5}$) particle concentrations calculated from TEOMs and the corresponding APS size fraction measured during April 11 - May 26, 2002. The secondary Y-axis presents the individual TEOM PM_{10} and $PM_{2.5}$ mass concentrations.

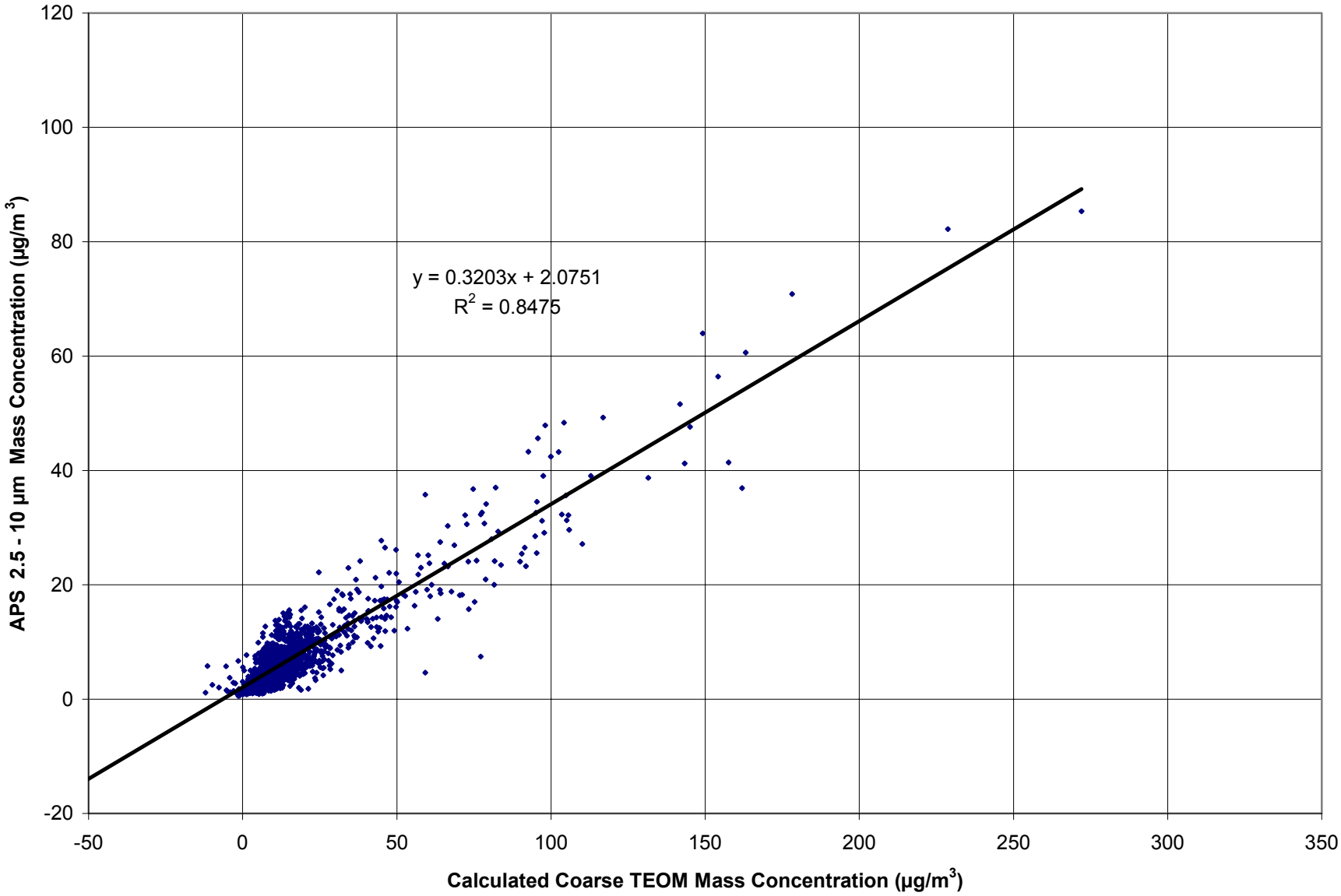

Figure 11. Scatter Plot comparison of coarse ($PM_{10} - PM_{2.5}$) particle concentrations calculated from TEOMs and the corresponding APS size fraction measured during quarter.